The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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LONG-TERM GOALS

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

For the current reporting period, our efforts have centered on the analysis of the data we gathered during the CARMA-IV field campaign. The goals of this campaign were as follows:

- Assemble a database on the size-resolved aerosol hygroscopicity for marine air off of the California coast

- Assess the relative importance of mixing of super-inversion air, and below cloud aerosol concentration on the cloud optical thickness and albedo. One particularly interesting facet of this assessment would be an appraisal of the impact of inhomogeneous mixing on the CDNC and cloud albedo (cf., Burnet and Brenguier, 2006). From this, assess the possibility of parameterizations that would predict the impact of these processes on the LWP and cloud base height in the MBL

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14. ABSTRACT

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

15. SUBJECT TERMS

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- Assess the importance of elemental carbon (EC) particles as CCN

- Evaluate the impact of chemistry on aerosol hygroscopicity during vertical transport in the MBL

APPROACH

Obtaining reliable measurements of both super-micron aerosol particles and their hygroscopicity is a major goal of our program. Our primary tool for this goal has been the AHS described in both the 2006 annual report and in Hegg et al (2007), coupled with the Descriptive Hygroscopic Growth Factor (DHGF) analysis technique also described in these documents. For the association of the measured hygroscopicity as a function of size with chemical composition, we have used several different substrate-based approaches for measuring the aerosol chemistry.

Assessing the relative importance of mixing of super-inversion air, and below cloud aerosol concentration on the cloud optical thickness and albedo was our second objective. In particular the impact of inhomogeneous mixing on the CDNC and cloud albedo using the approach of Burnet and Brenguier (2006) was to have been an important component of the analysis. From this, we then hoped to assess the possibility of parameterizations that would predict the impact of these processes on the LWP and cloud base height in the MBL.

The methodology for achieving the third objective involves the use of two instrument new to our program. The first of these is the annular geometry CCN spectrometer manufactured by DMT Inc. based on the design of Roberts and Nenes (2005). This will yield a continuous record of the CCN concentration at five supersaturations with a time resolution of about 10 minutes. This data will be compared with the concentration of EC bearing particles measured by the SP2 instrument manufactured by DMT Inc. and recently evaluated and described by Moteki and Kondo (2007). The comparison will yield the fraction of the CCN number concentration at each supersaturation that contain EC.

The final objective, evolution of the aerosol hygroscopicity in the vertical due to chemical activity has been examined by comparing both the hygroscopicity and chemistry of the aerosol at different altitudes in the MBL as per Hegg et al (2007).

WORK COMPLETED

The analysis of the data from the CARMA-IV campaign has been partially completed, with results pertinent to our first and fourth objectives listed above in hand. The third objective has been stalled due to problems with the analysis of the SP2 data. Essentially, the data volume from this device is so immense and complicated that ways of dealing with it have proven elusive. We will pursue this more aggressively in the coming year. The second objective, the analysis of the multiple linkages between cloud optical depth and albedo, and the proximate parameters that control them, has required some adjustments on our part. The main thrust of our analysis was to have been the examination of the impact of inhomogeneous mixing on cloud albedo as per Burnet and Brenguier (2006). However, our analysis of 21 vertical profiles through the California stratocumulus, using the methodology of Burnet and Brenguier, has yielded only one possible instance of such mixing. Hence, it is our current view that this process is so rare that it has no significance, at least for our venue. Consequently, we will be

re-directing our analysis in the upcoming year to address the issue of the relative impact of conventional mixing and aerosols on stratocumulus albedo.

RESULTS

The DHGF spectra obtained in CARMA-IV were, in general, quite similar to those we observed in CARMA-III. The average of marine spectra sampled at 30 m MSL and in the range 100-500 m MSL from CARMA-IV are shown in Figure 1.

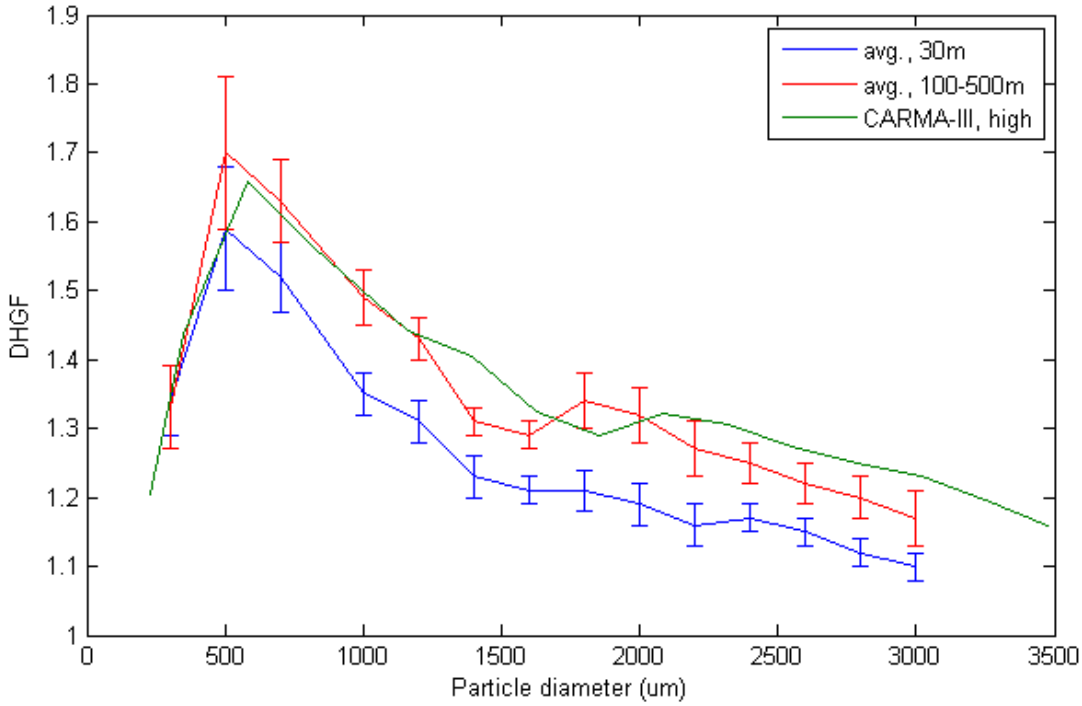

Figure 1. DHGF spectra measured during CARMA-IV for two different altitudes. For comparison, the mean spectrum from CARMA-III for the higher of the two altitude intervals is also shown. All the spectra show relatively low hygroscopicity at larger sizes but there is a significant difference in the mean spectra with altitude with the higher altitudes showing more hygroscopicity.

The spectra for both campaigns show substantially reduced hygroscopicity at the supermicron sizes, as we have noted in already published studies (Hegg et al, 2006; 2007). From the standpoint of our fourth objective, the more telling characteristic is the significant increase in hygroscopicity with altitude, most evident in the CARMA-III data. The chemical composition of the aerosol associated with this change is most interesting and is shown in Figure 2. Sulfate and nitrate concentrations are similar at the two

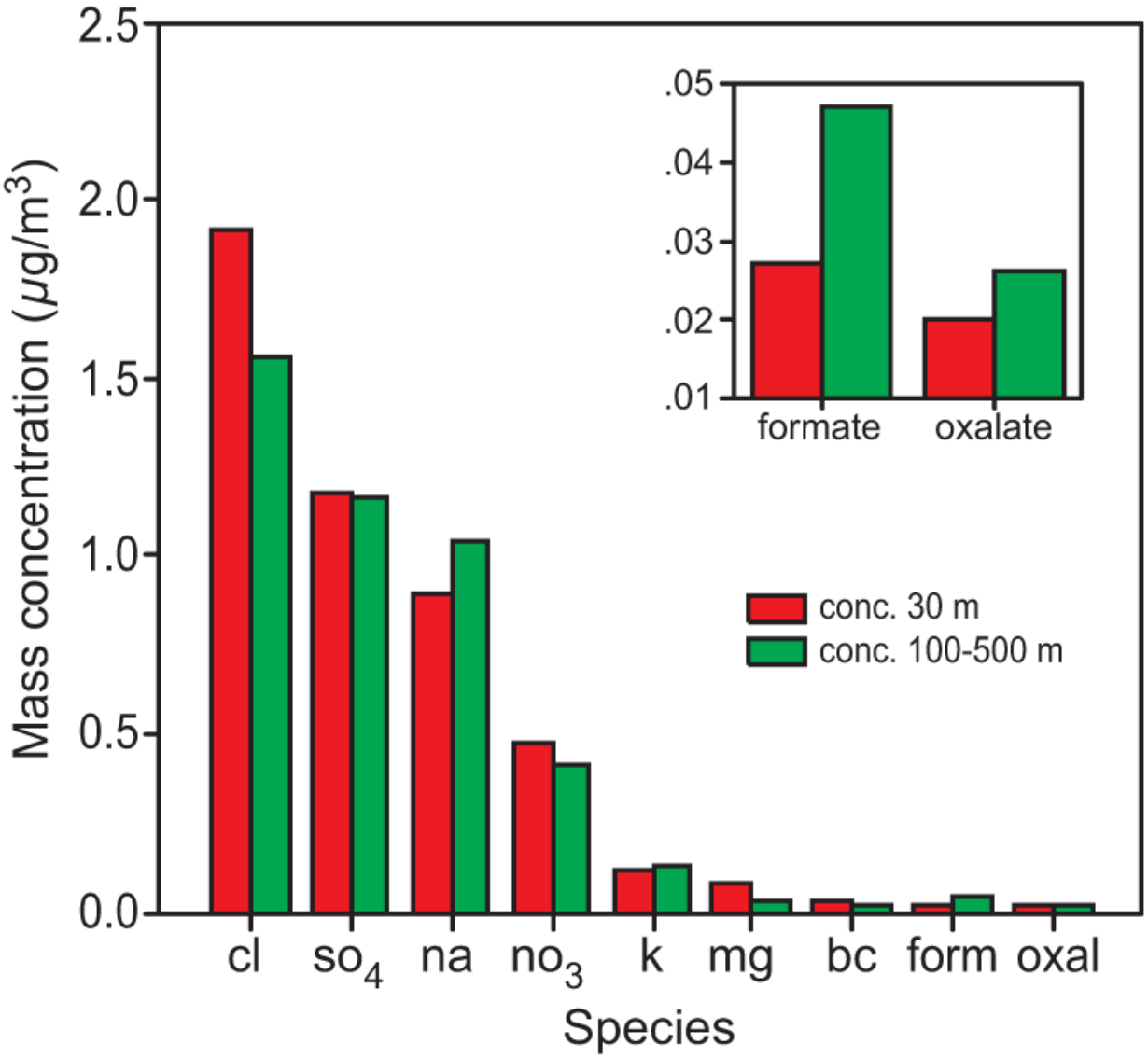

Figure 2. Comparison of the mean chemical concentration of the dominant soluble chemical constituents of the aerosol at high (>160 m) and low altitude (30 m) during the study period. The associated DHGF spectra are shown in Figure 1. The insert shows more clearly the changes in formate and oxalate

altitudes, suggesting little condensation of soluble secondary inorganics. The NaCl concentrations, as might be expected, are somewhat lower aloft. Hence, the increase in hygroscopicity cannot be explained by the simple mechanism of an increase in the soluble inorganic mass fraction (note that the total mass is essentially the same at the two altitudes). On the other hand, as shown by the insert, the formate and oxalate concentrations are appreciably higher aloft. Since these are well known oxidation products of long chain alkanoic acids and alcohols, which are know surfactants on marine aerosol, we interpret this as evidence that the increase in hygroscopicity with altitude is due to the in situ oxidation of surface films as the aerosol is transported upward in the boundary layer. A more detailed discussion of this is in Hegg et al, 2008.

Another main thrust of our analysis has been the source attribution of the aerosols, and their hygroscopicity, for our coastal venue. To this end, we have employed a well-known receptor model,

the US EPA UNMIX 2.3 model, to resolve the aerosol sources for our measured aerosol. Application of this model to our chemical composition data set lead to the resolution of three main aerosol sources for CARMA-IV, namely, biomass burning, pollution and marine. To illustrate the variability in the contribution of these sources to individual samples, the mass fractions for each aerosol type/source for each of the chemical samples obtained in CARMA-IV are shown in Figure 3.

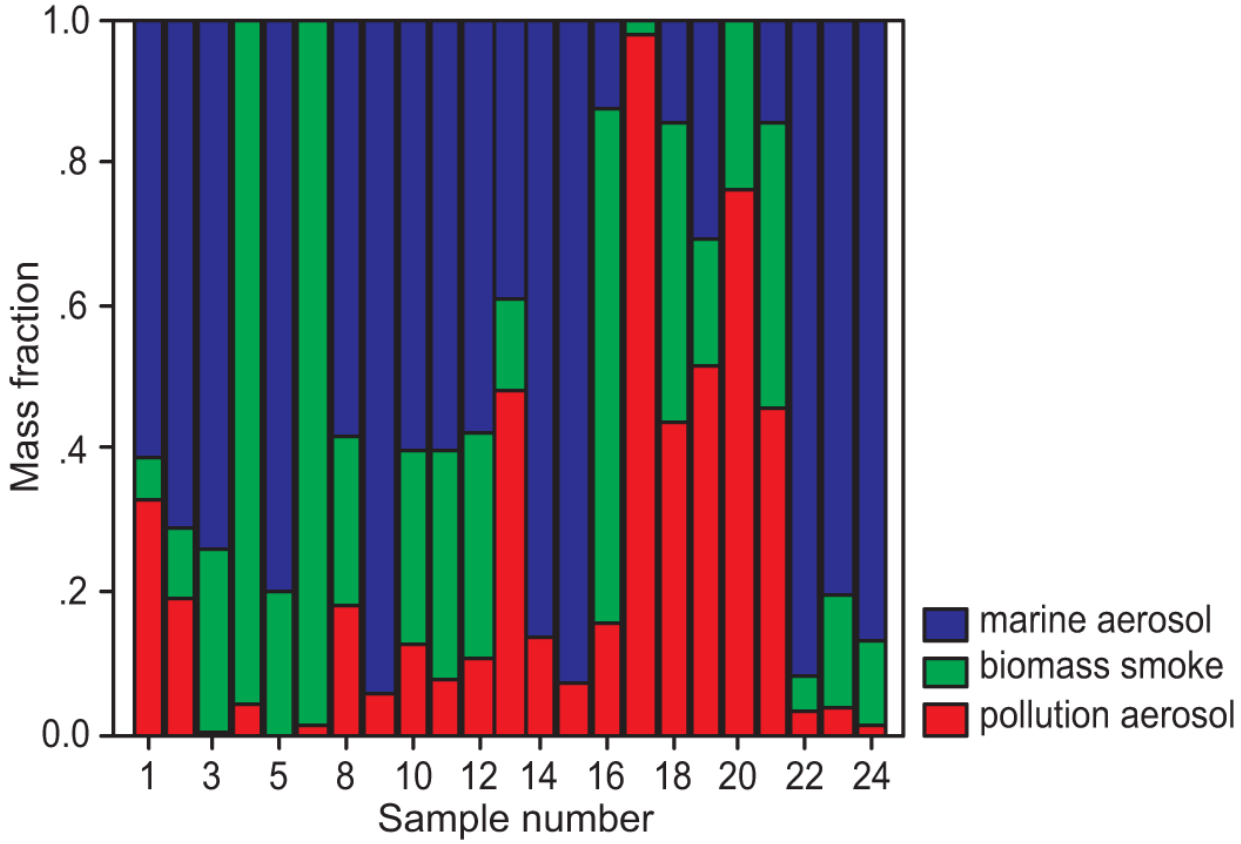

Figure 3. Mass fractions of each of the three aerosol types in the study area for each of the aerosol samples analyzed. For most samples, as expected, the marine source dominates but there are clear instances of both pollution and biomass aerosols controlling the composition of the aerosol.

Given these three distinct aerosol types, an important issue is how to predict the hygroscopicity of mixtures of the three shown in Figure 3 if one knows the typical hygroscopicity of the individual types. One such methodology is the Zdanovskii, Stokes and Robinson (ZSR) mixing rule, which has been used for growth factors (essentially DHGF's) for pure chemical species derived from laboratory studies (e.g., Gysel et al, 2006). We now apply the ZSR rule to the mean DHGF's associated with our aerosol types to predict the DHGF's of the internal mixtures represented in Figure 3. A comparison of the predictions of the mixing rule with the actual measured DHGF's has suggested that the rule has substantial prognostic power for the submicron size range but little for the supermicron. Reasons for this, together with more details on the overall analysis are given in Hegg et al, 2008.

IMPACT/APPLICATIONS

The results from CARMA-IV support those obtained earlier as to the shape of DHGF size spectra, and the differences seen between different aerosol types. The size dependence in particular will be quite valuable in refining estimates of direct aerosol forcing in the marine boundary layer. The variations of

hygroscopicity with altitude should be similarly valuable. The prognostic power of the ZSR mixing rule in deriving the overall hygroscopicity of submicron aerosol based on the aerosol sources should prove valuable in refining numerical transport model calculations.

TRANSITIONS

None.

RELATED PROJECTS

The size dependent hygroscopic growth of aerosols, including super-micron aerosols (cf., Quinn et al, 1998), is a major factor in both the radiative energy balance of the lower marine atmosphere and the propagation of radiation through the MBL. Such radiation properties are necessary parameters for numerical modelers developing prognostic models. It is, furthermore, an aerosol characteristic closely related to CCN activity and, indeed, such activity can be predicted from it. Hence, these measurements are highly relevant to determination of CCN spectra and thus of the microphysics of MBL clouds. Finally, numerical transport models now incorporate aerosol sources and can predict the amount of aerosol from the incorporated sources present at any particular point in the model domain. The prognostic power of the ZSR mixing rule should permit the internal calculation of the hygroscopicity of the modeled aerosols as well.

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PUBLICATIONS

Hegg, D.A., D.S. Covert and H.H. Jonsson, Measurements of size-resolved hygroscopicity in the California coastal zone, Atmos. Chem. Phys. Discuss., 8, 10531-10560, 2008.

PATENTS

None.